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PB-298 519

Factors Affecting Dry Deposition of SO₂ on Forests and Grasslands

(U.S.) Environmental Sciences Research Lab, Research Triangle Park, NC

1 Dec 77

TECHNICAL REPORT DATA (Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA-600/J-78-117	2. JOURNAL ARTICLE	3. RE(PB 298519
4. TITLE AND SUBTITLE FACTORS AFFECTING DRY DEPOSITION OF SO ₂ ON FORESTS AND GRASSLANDS		5. REPORT DATE
7. AUTHOR(S) J.H. Shreffler		6. PERFORMING ORGANIZATION CODE
9. PERFORMING ORGANIZATION NAME AND ADDRESS Same as Box 12		8. PERFORMING ORGANIZATION REPORT NO.
12. SPONSORING AGENCY NAME AND ADDRESS Environmental Sciences Research Laboratory - RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711		10. PROGRAM ELEMENT NO. 1AA603 AB-28 (FY78)
15. SUPPLEMENTARY NOTES REFERENCE: Atmos. Environ., 12(6-7):1497-1503, 1978.		11. CONTRACT/GRANT NO.
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		14. SPONSORING AGENCY CODE EPA/600/09
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
*Air pollution *Sulfur dioxide *Deposition *Forestry *Grassland		13B 07B 02F 08F
18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS (This Report) UNCLASSIFIED	21. NO. OF PAGES 9
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE PC402/MEROL

EPA Form 2220-1 (9-73)

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SPRINGFIELD, VA. 22161

FACTORS AFFECTING DRY DEPOSITION OF SO_2 ON FORESTS AND GRASSLANDS

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(First received 30 August 1977 and in final form 1 December 1977)

Abstract—Deposition velocities for SO_2 over forests and grasslands are derived through a mass conservation approach utilizing established empirical relations descriptive of the atmospheric transport of a gaseous contaminant above and within a vegetational canopy. Of particular interest are situations where surface resistance to uptake is relatively small and does not overwhelm atmospheric transport in the determination of deposition. To this end, emphasis is placed on cases assuming the vegetation surface is a perfect sink or has resistance approximating minimum stomatal resistance.

This study indicates that deposition velocity, v_d , is relatively insensitive to canopy height, shape, roughness length, and extinction coefficients, but v_d is relatively sensitive to atmospheric stability, zero-plane displacement, surface resistance, leaf area index, and friction velocity. It follows that the latter parameters should be reported to allow for meaningful comparisons between v_d 's determined under various experimental conditions. Using reasonable parameter choices, predictions by this method agree well with previous observations, yielding v_d in the neighborhood of 1 cm s^{-1} over grass (or other low-set vegetation) and a value of 2 to 3 times as great over a forest. Only if a forest canopy could be considered a perfect sink would v_d 's approaching 10 cm s^{-1} be expected.

NOMENCLATURE

$A(z)$	leaf area density
a_1	canopy extinction coefficient for $K(z)$
a_2	canopy extinction coefficient for $U(z)$
$c(z)$	pollutant concentration, mass/volume
d	zero-plane displacement
D	molecular diffusivity of pollutant in air
f	factor for varying the ratio $LAI/(r_a + r_s)$
h	canopy height
$I(z)$	interfacial flux, flux at leaf surface
k	von Karman constant
$K(z)$	vertical eddy diffusivity
L	Monin-Obukhov length
LAI	leaf area index
r	reference height for r_s
r_a	aerodynamic resistance
r_s	surface resistance
u	friction velocity
$U(z)$	wind speed
v_d	deposition velocity
z	vertical distance from the ground
z_0	roughness length
$\eta(z)$	$(z - d)/L$
l	length scale for the width of a leaf
ν	kinematic viscosity of air
ϕ_M	non-dimensional wind shear
ϕ_H	non-dimensional temperature gradient
$\psi_{M,H}$	defined by Equation (3)

INTRODUCTION

As a specification of the potential for dry deposition of SO_2 , the deposition velocity, v_d , is a convenient expression for use in long-range transport models

(Scriven and Fisher, 1975; Bolin and Persson, 1975). The flux of the pollutant into the surface is computed simply as the product of v_d and the predicted concentration (mass/volume) at a convenient reference height near the surface.

Generalizing the results of experiments on atmospheric transport of sensible heat, water vapor and momentum, Hicks and Liss (1976) have presented a method for predicting v_d of SO_2 (or other reactive gases) over lakes and oceans. Such predictions would be of importance in computing global budgets of sulphur and for use in long-range transport models, particularly in European situations where moderate expanses of open water may separate the source and receptor locations. It is recognized, of course, that vegetated land surfaces also represent a sink for SO_2 (Hill and Chamberlain, 1976), and it would be advantageous to deduce representative values of v_d for these surfaces also. However, this is a more involved task than prediction for oceanic surfaces. Besides the factors affecting v_d which were cited by Hicks and Liss, including wind speed and atmospheric stability, additional complications are introduced by canopy morphology and physiology. There would be several factors tending to enhance deposition on a canopy when compared to a flat surface, among them, the relatively large foliage area, greater turbulent mixing, and exposure of the leaves to comparatively high mean wind speeds which lower aerodynamic resistance. On the other hand, if the pollutant gas is removed, not on the leaf cuticle, but only after diffusion through open stomata to the interior of the leaf, a significant surface resistance is encountered which has no counterpart in

* On assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce.

the open water surface. Furthermore, vertical turbulent transfer tends to decrease rapidly downward in a canopy, preventing foliage at lower levels from reaching its potential as a sink.

Measurements of deposition velocity have generally been restricted to short plant canopies or water (Owers and Powell, 1974; Shepherd, 1974; Whelpdale and Shaw, 1974), although a recent attempt has been made to infer v_d for SO_2 over a forest (Petit *et al.*, 1976). An interesting result from these studies is that v_d appears to be of similar magnitudes for water and vegetation, despite the disparity in the surface areas involved. Further studies in forested areas would be useful, but the deduction of fluxes by the profile method is usually difficult and expensive while the direct measurement of deposition by use of S^{35} tracer is probably not feasible on large scales. As an alternative, insight into deposition on various canopies might also be gained by synthesizing the wealth of empirical knowledge regarding atmospheric transport, canopy structure and leaf physiology. To this end, a theoretical treatment, describing the atmospheric transfer of gaseous pollutants to a surface of fibrous character, was presented by Shreffler (1976) and was shown to be consistent with the Chamberlain (1966) data taken over an artificial grass-like surface. It is the intent here to modify and extend that treatment to simulate natural surfaces, forests and grasslands, and to systematically examine the factors affecting the magnitude of deposition velocity over such surfaces.

MASS CONSERVATION APPLIED TO CANOPY DEPOSITION

The theoretical treatment is one-dimensional and, therefore, is strictly applicable only under conditions approaching horizontal homogeneity. The basic equation derived here is a statement of mass conservation for a gaseous pollutant which penetrates a vegetational canopy from above and is subject to removal on or within the leaves. The transfer of the pollutant from the free atmosphere to the ultimate sink is divided into four components: turbulent transfer above the canopy, within the canopy, interfacial transfer (that dominated by molecular diffusion very near the leaf surface) and finally (if applicable), diffusion through open stomata to the interior of the leaf.

Given a plant canopy of height h and zero-plane displacement d , an atmospheric surface layer is assumed above h with turbulent transfer coefficient

$$K(z) = u_* k(z - d) / \phi_H(\eta) \quad (1)$$

where $k = 0.41$, u_* is the friction velocity, and z is the distance from ground level. The expressions for non-dimensional wind shear, ϕ_M , and temperature gradient, ϕ_H , above the canopy follow the suggestions of Dyer (1974),

$$\begin{aligned} \phi_M(\eta) &= (1 - 16\eta)^{-1/4} & \eta < 0 \\ \phi_H(\eta) &= (1 - 16\eta)^{-1/2} & \eta < 0 \end{aligned} \quad (2)$$

$$\phi_M(\eta) = \phi_H(\eta) = 1 + 5\eta \quad \eta > 0$$

where $\eta = \eta(z) = (z - d)/L$ and L is the Monin-Obukov length scale. Stability functions are defined as

$$\psi_j(\eta(z_1), \eta(z_2)) = \int_{\eta(z_1)}^{\eta(z_2)} \frac{1 - \phi_j(\zeta)}{\zeta} d\zeta \quad (3)$$

where j is either M or H . The method for closed integration of (3) under unstable conditions is given by Paulson (1970). As assumed by Hicks and Liss (1976), the transfer of the pollutant will be considered similar to that of sensible heat rather than momentum.

Within the canopy, $z < h$, an exponentially decreasing form (Cionco, 1972) is assumed for the turbulent transfer coefficient.

$$K(z) = K(h) \exp(-a_1(1 - z/h)) \quad (4)$$

The flux of the gas into a leaf is expressed in terms of an aerodynamic resistance, r_a , and surface resistance, r_s , giving

$$I(z) = c(z)/(r_a(z) + r_s) \quad (5)$$

where $c(z)$ is the concentration at level z within the canopy. The surface resistance, which might also be a function of z , is defined here on a leaf area basis and not as a bulk resistance. If the outer leaf surface is a perfect sink, $r_s = 0$; if the ultimate sink is in the leaf interior, r_s must at least be equal to stomatal resistance and may include other internal resistances (Bennett *et al.*, 1973). Stomatal resistance is dependent on various environmental factors and becomes virtually infinite at night (Meidner and Mansfield, 1968). Measurement of mass transfer to and from isolated artificial leaves by Thom (1968) and Chamberlain (1974) indicate that r_s may be approximated by

$$r_s(z) = 1.4(\ell/vU(z))^{1/2} (v/D)^{2/3} \quad (6)$$

where ℓ is a representative leaf dimension, U is the mean wind speed, and D is the molecular diffusivity of the pollutant in air. It should be noted that Thom's (1968) expression is half as large as (6) since he defined r_s based on the flux from both sides of the leaf but the area of one side. Some caveats are in order concerning application of (6); in particular, fluttering of natural leaves will tend to decrease r_s (Schuepp, 1972) while interference from neighboring leaves may tend to increase r_s . Further, there is no sound basis for applying (6) when ℓ is much outside the range used in the experiments, $\ell = 4$ cm for Thom and $\ell = 4$ and 11 cm for Chamberlain. Specifically, it would be inadvisable to apply (6) to the needles of a conifer, although other data are available (Jarvis *et al.*, 1976).

The expression for the mean wind speed above the canopy, $z > h$, is given by

$$U(z) = (u_*/k) [1n((z - d)/z_0) - \psi_M(\eta(d + z_0), \eta(z))] \quad (7)$$

where z_0 is the roughness length. Within the canopy, wind speed drops off exponentially (Cionco, 1972) as

$$U(z) = U(h) \exp(-a_2(1 - z/h)), \quad (8)$$

and it is this speed that is used in (6) for computing r_a .

Between h and a reference level, $r > h$, a constant downward flux F is assumed. As the pollutant penetrates the canopy, it is subject to deposition and $F(z)$ decreases downward. Letting $A(z)$ be the leaf area per unit volume, the mass conservation equation is

$$F(h) - \int_z^h f \cdot A(z) \cdot I(z) dz = K(z) c'(z), \quad (9)$$

where primes indicate differentiation with respect to z , and f is a multiplicative factor (considered unity here) which will be used in later discussion to investigate the effects of various model parameters. Differentiating (9) results in

$$c''(z) + \frac{K'(z)}{K(z)} c'(z) - \frac{f A(z)}{(r_a(z) + r_s) K(z)} c(z) = 0. \quad (10)$$

The mass conservation equation (10) is solved as a two point boundary value problem (Keller, 1968) using a seventh order predictor-corrector. The vegetation substrate is assumed impervious so that

$$c'(0) = 0. \quad (11)$$

Other forms of the lower boundary condition could be enforced to describe uptake by the soil (Shreffler, 1976), but in this paper emphasis will be placed on deposition to the canopy proper. The upper boundary condition, applied at h , follows from the flux-gradient relation associated with (1) and (2).

$$c(h) + \frac{(h-d)}{\phi_H(\eta(h))} \left[\ln \left(\frac{r-d}{h-d} \right) - \psi_H(\eta(h), \eta(r)) \right] c'(h) = c(r). \quad (12)$$

Solution of (10) subject to (11) and (12) requires specification of the canopy morphology and physiology (h , d , z_0 , $A(z)$ and r_s) and atmospheric conditions (u_* and L). If the concentration is normalized by $c(r)$, the flux above h is then the deposition velocity at level r .

DEFINITION OF STANDARD CASES

In this section, selections are made for values of parameters describing the canopy structure and atmospheric conditions with the intention of defining standard cases which are typical or reasonable. The results from these standard cases will be used as a basis for evaluating the impact of parameter choices on the predicted deposition velocities.

A leaf area density profile, Fig. 1, representing a deciduous forest canopy with sparse grass undergrowth is adapted from Rauner (1976) and is taken to describe the standard forest canopy. For this study, the standard grass canopy is described by a leaf area density five times as great as the sparse grass in Fig. 1. The standard canopies have height of 16 m and 0.6 m,

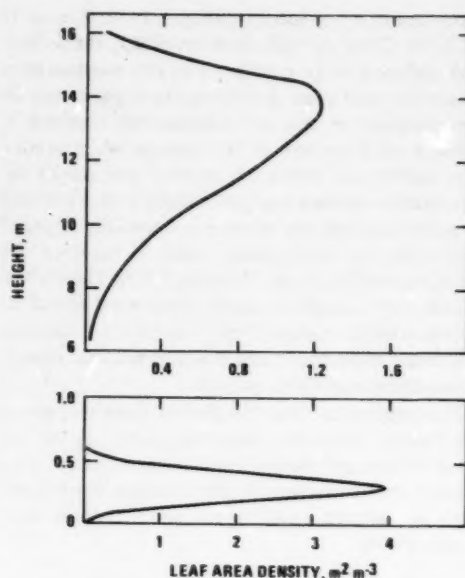


Fig. 1. Leaf area density, both sides of the leaf, for a deciduous forest canopy (upper) with sparse grass undergrowth (lower). Adapted from Rauner (1976).

with leaf area indices (LAI , based on the area of both sides of the leaf) of 6.6 and 5.0, respectively. The forest canopy is typical of both coniferous and deciduous forests where LAI 's typically range from 2 to 15 (Jarvis *et al.*, 1976; Rauner, 1976). The grass canopy is intended to represent any low-set vegetation, including a field crop.

In addition, the following expressions and parameter values are used to delineate the standard cases.

$$\begin{aligned} d &= 0.75h \\ z_0 &= 0.36(h-d) \\ r &= d + 10 \text{ m} \\ a_1 &= 2.0 \text{ (forest)} \\ a_2 &= 1.0 \\ a_1 &= 3.0 \text{ (grass)} \\ a_2 &= 2.5 \\ \ell &= 2 \text{ cm} \\ r_s &= 0 \text{ or } 1 \text{ s cm}^{-1} \\ D &= 0.122 \text{ cm}^2 \text{ s}^{-1} \\ u_* &= 40 \text{ cm s}^{-1} \\ L &= \infty \end{aligned}$$

The expressions for d and z_0 are adopted from wind tunnel data of Thom (1971) and agree reasonably well with forest data also (Rauner, 1976; Jarvis *et al.*, 1976). The extinction coefficients, a_1 and a_2 , for the eddy diffusivity and mean wind speed follow the trend of values summarized by Cionco (1972). Low, dense canopies tend to have higher extinction coefficients. The leaf dimension, ℓ , is the scale used in computing the molecularly dominated aerodynamic resistance by (6). Surface resistance is taken either as zero, implying that the entire leaf surface is a perfect sink, or as 1 s cm^{-1} , approximating a lower bound for stomatal

resistance in many species (Saugier, 1976; Rauner *et al.*, 1976). Given the high solubility of SO_2 , the perfect-sink surface may be envisioned as one overlain by a water film, and there is evidence to suggest that an accumulation of dew can substantially diminish r_s (Fowler and Unsworth, 1974). Likewise, water interior to a healthy leaf with a dry exterior may render the mesophyll resistance negligible (Kabel *et al.*, 1976) and so result in a situation where r_s is essentially stomatal resistance. An appropriate value is adopted for diffusivity of SO_2 in air (Andrew, 1955). The friction velocity and roughness imply mean wind speeds at reference height of about 5 m s^{-1} and 2 m s^{-1} for grass and forest respectively, and it is assumed that neutral atmospheric conditions prevail.

It is emphasized that the choices presented above are merely justifiable when compared to relevant observations, and the results will be presented in such a manner that the impact of other reasonable selections may be inferred qualitatively and, in some case, quantitatively.

DISCUSSION OF RESULTS

In all cases discussed, the standard values of model parameters apply unless otherwise specifically stated. Figure 2 presents predicted v_d as a function of u_* for forest and grass canopies with $r_s = 0$ and 1 s cm^{-1} ; for comparative purposes, Fig. 2 also shows v_d predicted by a method of Hicks and Liss (1976) over an ocean surface. To a first approximation, surfaces with $r_s = 0$, including the ocean, show a linear increase in v_d with u_* . Introduction of a surface resistance will limit the increase. If r_s remains small, v_d over a forest can

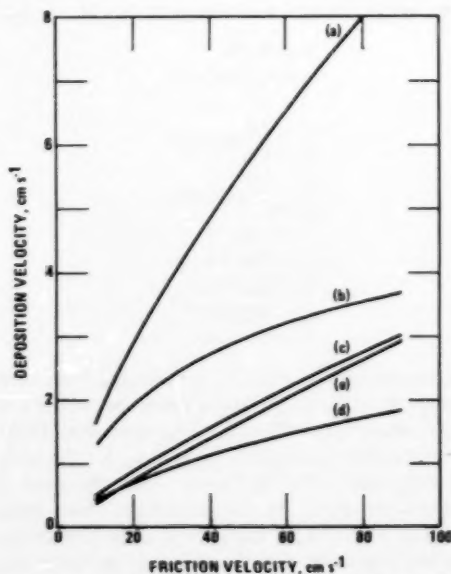


Fig. 2. Predicted v_d as a function of u_* over the forest canopy with $r_s = 0$ (a) and $r_s = 1 \text{ s cm}^{-1}$ (b); over the grass canopy with $r_s = 0$ (c) and $r_s = 1 \text{ s cm}^{-1}$ (d); over an ocean surface (e).

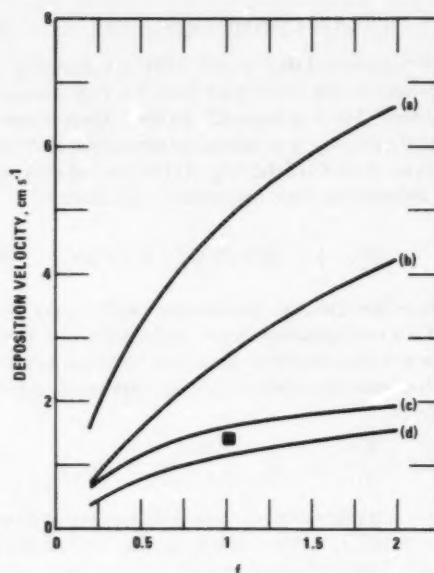


Fig. 3. Predicted v_d as a function of f , a multiplicative factor of $LAI/(r_a + r_s)$ for the standard cases. Curves are labeled as in Fig. 2, and the ocean value is indicated by ■.

significantly exceed that over a flat water surface. Figure 3 depicts the effect of changing the factor f as previously defined in (9). This factor may be precisely interpreted as a multiplier of LAI ; also, f may be used to assess the impact of changing $(r_a + r_s)$, although this relation is not precise since r_a , but not r_s , is taken as a function of z . For instance, in the upper portion of the forest canopy, $r_a \approx 0.67 \text{ s cm}^{-1}$. Increasing r_s from 1 to 2.67 s cm^{-1} would then about double $(r_a + r_s)$, and, in Fig. 3, $f = 0.5$ would be the appropriate factor to apply to curve (b) (or $f = 0.2$ to curve (a)). Although Fig. 3 strictly applies only for $u_* = 40 \text{ cm s}^{-1}$, estimates for other u_* are possible simply by computing f based on a corrected r_a and making a proportional adjustment to the resulting v_d based on Fig. 2.

In explicitly treating the vertical structure of the canopy, the method discussed in this paper differs fundamentally from those which consider that all layers of foliage participate equally in the removal process (e.g. Murphy, 1976). The effective shielding of low level foliage may be seen in Fig. 2 by comparing results for the perfect sink canopies, curves (a) and (c) with those for the ocean, curve (e). Over the grass canopy, v_d is only slightly larger than over the ocean surface, although the grass offers an area five times as great. Values of v_d for the forest canopy are substantially higher but are still not commensurate with the available surface area ($LAI = 6.6$).

To further evaluate the effects of model assumptions on the predicted v_d , the principal parameters were varied individually while holding all others at the standard values, and the degree of variation reflects that which might be expected based on observational studies. Results of the parameter variation are pre-

Table 1. Percentage changes in the predicted deposition velocity resulting from variation of basic parameters describing the grass canopy. For the standard case predictions, refer to Fig. 3, curves (c) and (d), $f = 1$

Parameter	Parameter value	% change from standard case, $r_s = 0 \text{ s cm}^{-1}$	% change from standard case, $r_s = 1 \text{ s cm}^{-1}$
z_0	$0.4(h-d)$	-2	-2
	$0.2(h-d)$	8	5
a_1, a_2	4.0, 4.0	-16	-14
	2.0, 2.0	10	8
h	2 m	13	9
	0.2 m	-10	-7
Canopy shape	Uniform	15	7
L	-40 m	10	8
	-10 m	23	16
	40 m	-11	-9
	10 m	-32	-27
d	$0.85h$	-17	-15
	$0.55h$	20	16
r	$d + 1 \text{ m}$	28	19
	$d + 5 \text{ m}$	7	5

Table 2. Percentage changes in the predicted deposition velocity resulting from variation of basic parameters describing the forest canopy. For the standard case predictions, refer to Fig. 3, curves (a) and (b), $f = 1$

Parameter	Parameter value	% change from standard case, $r_s = 0 \text{ s cm}^{-1}$	% change from standard case, $r_s = 1 \text{ s cm}^{-1}$
z_0	$0.4(h-d)$	-3	-1
	$0.2(h-d)$	12	6
a_1, a_2	3.0, 1.5	-5	-5
	1.0, 0.5	6	4
h	32 m	26	13
	8 m	-17	-10
Canopy shape	Uniform	-20	-15
L	-40 m	21	11
	-10 m	29	17
	40 m	-20	-14
	10 m	-51	-40
d	$0.85h$	-22	-15
	$0.55h$	37	19
r	$d + 5 \text{ m}$	26	13
	$d + 20 \text{ m}$	-17	-10

sented in Tables 1 and 2 for the grass and forest canopies, respectively. Some parameter choices do not appear to be especially critical, such as those for z_0 , a_1 and a_2 . Canopy height (with the same LAI and general shape) has a stronger influence, but only when varied considerably, and it should be possible to specify h much more closely than in this example. To test the influence of canopy shape as given in Fig. 1, computations were also carried out using a uniform canopy with the same LAI. The shape seems to be of some importance, with canopies having greater area in the upper levels giving the largest predicted v_d 's. Atmospheric stability, as expressed by L , is quite important, especially in its ability to suppress de-

position under stable conditions. Also of great importance is the choice for zero-plane displacement which has a major impact on the magnitude of K both above and within the canopy; taking d from $0.85h$ to $0.55h$ involves a threefold increase in K . Although the reference height should be known accurately, the dependence on r is included to emphasize the need for its specification when reporting v_d .

Each of the parameters examined in Tables 1 and 2, and including u_* in Fig. 2, deal in some manner with the atmospheric transfer of the pollutant. As the surface resistance increases, the changes in assumptions concerning the atmospheric phase of transport become less significant; this can be seen in going from

$r_s = 0$ to $r_s = 1 \text{ s cm}^{-1}$. The latter resistance was chosen as representing minimum stomatal resistance and certainly does not dominate the atmospheric transport. However, in general it is true that

$$v_g < LAI/r_s \quad (13)$$

and $v_g \rightarrow LAI/r_s$ as $(LAI/r_s) \rightarrow 0$.

So, as r_s increases, or the canopy becomes sparse, v_g will be only slightly less than LAI/r_s . What constitutes a large r_s conducive to this approximation is relative. If the atmosphere is unstable with high u_g , then r_s need not be very large to have a notable impact but, of course, under any circumstances the effect of atmospheric transfer is never completely lost. For the standard forest with $r_s = 6.6 \text{ s cm}^{-1}$ ($LAI/r_s = 1 \text{ cm s}^{-1}$), the predicted $v_g = 0.82 \text{ cm s}^{-1}$; with $r_s = 13.2 \text{ s cm}^{-1}$, the result is $v_g = 0.45 \text{ cm s}^{-1}$. As r_s increases, v_g must inevitably be forced downward leaving tall and short canopies of similar LAI 's with similar v_g 's. This effect may be seen in the left of Fig. 3 where the grass and forest predictions converge.

Although a number of experiments have been performed to determine v_g for grass (Garland *et al.*, 1973; Shepherd, 1974; Owers and Powell, 1974; Whelpdale and Shaw, 1974), the reporting of the experiments has, to various degrees, not included basic information concerning the state of the atmosphere and structure of the canopy, most notably the LAI . For this reason exact comparisons with the model are not possible. However, the first three papers cited above find typical values of v_g in the neighborhood of 0.8 cm s^{-1} . This is comparable to a model prediction using the reasonable values of $h = 10 \text{ cm}$, $LAI = 3$, $u_g = 30 \text{ cm s}^{-1}$, $r_s = 1 \text{ s cm}^{-1}$ and $r = 50 \text{ cm}$. The values of v_g reported by Whelpdale and Shaw are significantly higher, but are defined in a non-standard manner involving an extrapolated surface concentration. However, their data do confirm that stable conditions can effect large relative reduction in v_g . Recent measurements by Petit *et al.* (1976) over a forest lead to v_g 's of 1.8 and 3.7 cm s^{-1} . The forest was roughly comparable to that described by the standard case, so these determined v_g seem consistent with model predictions, again assuming a surface resistance of about 1 s cm^{-1} .

CONCLUSIONS

Predictions of SO_2 deposition velocity have been made by solving a mass conservation equation descriptive of transport and removal of a gaseous pollutant in a vegetational canopy. The equation formulation relied heavily on observations of the diabatic surface layer, canopy flow and vegetation structure. Of particular interest in this paper was the case in which surface resistance does not dominate the atmospheric phase of transport in the final determination of v_g .

It should not be inferred that precise values of v_g may be derived by this method. The model involves idealizing assumptions unlikely to be fully satisfied, even on a

carefully chosen experimental site. Moreover, it is not expected that all parameters would be known or measured accurately at any given time. However, a systematic examination of all factors involved in the deposition process facilitates the interpretation of existing observational results and sets general levels for v_g to be expected under different circumstances.

Assuming $r_s = 1 \text{ s cm}^{-1}$, a value taken as representative of minimum stomatal resistance, the model predicts values of v_g which compare well with measured values over short grass and a forest. Both observations and predictions indicate that v_g over a forest will be 2-3 times as great as over grass. Furthermore, it may be possible under ordinary conditions for v_g over a forest to be twice as large as over the ocean.

Obviously it is important to arrive at realistic values of r_s , including diurnal and seasonal variations. Because of the high solubility of SO_2 , it is of particular interest whether or not moist vegetation may approximate a perfect sink surface, $r_s = 0$. Model predictions indicate that v_g 's approaching 10 cm s^{-1} may not be uncommon in such a case. On the other hand, when r_s exceeds 10 s cm^{-1} , atmospheric transport becomes relatively unimportant and v_g may be approximated by LAI/r_s .

The parameter analysis performed with the model points out the special need in future experimental studies to report r , h , d , L , LAI and u_g . Only by knowing these parameters can there be a basis for meaningful comparisons of v_g 's determined under different conditions.

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